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Research article

Odor impact assessment of trace sulfur compounds from working faces of landfills in Beijing, China

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ABSTRACT

Odor pollution from landfills is causing a growing number of public complaints and concerns. Compared with hydrogen sulfide (H_2S) and ammonia (NH_3), odor impacts of trace sulfur compounds (TSCs) are arousing concerns due to their low odor threshold values (OTVs). Working face on landfill sites has been claimed as major source of odor impacts. This study estimated the odor impacts of fugitive TSCs from the working face of a large typical municipal solid waste (MSW) landfill in Beijing, China. A modified wind tunnel system was introduced to estimate emission rates of TSCs, which is a basic requirement for odor impact assessment. The odor activity value (OAV) method was introduced for odor evaluation. Fieldwork in the selected landfill was conducted from 2014 to 2015. Methyl mercaptan (CH_3SH), dimethyl sulfide, dimethyl disulfide (DMDS), and carbon disulfide (CS_2) were the TSCs studied in this work. The spatial concentration distributions of the TSCs were calculated on the basis of the Gaussian dispersion model in a "normal case" scenario and a "worst case" scenario. DMDS showed the highest emission rate (7.18 $\mu g\, m^{-2}\, s^{-1}$), and CH_3SH was the dominant odorous compound with an average emission rate of 4.58 $\mu g\, m^{-2}\, s^{-1}$. The dispersion modeling indicated that the odor impact distances of the TSCs in the studied landfill for the normal case and worst case scenarios were 495 ± 96 m and 9230 m at the downwind regions, respectively. Results of this study can benefit the formulation of strategies for odor control and abatement in landfill sites.

1. Introduction

Landfills have become the major approach for municipal solid waste (MSW) disposal in China, with over 60% of MSWs disposed in landfills (Beijing Solid Waste Administration Department, 2013; Zhang et al., 2010). Odor-related complaints in landfill sites have long been an issue, and the required engineering controls for limiting air pollution from MSW landfills have been adopted to protect the environment and nearby communities. Methane (CH_4) and carbon dioxide (CO_2) are the major components of landfill gas (LFG), comprising approximately 99% (V/V) of the total LFG; however, neither gas is odorous (Allen et al., 1997; McKendry, 2002). Non methane organic compounds (NMOCs), which comprise < 1% (V/V) of the total LFG, are considered the major source of odor pollution in landfills (Fang et al., 2012). Generally, unpleasant odors in landfill sites are associated with hydrogen sulfide (H_2S) and ammonia (NH_3), however, some organic sulfur compounds are also significant odorants, primarily mercaptans and sulfides (Saral et al., 2009), due to their low odor threshold values (OTVs), and they were defined as such as trace sulfur compounds (TSCs) here. For example, the OTV of methyl mercaptan (CH_3SH) is 0.15 $\mu g\, m^{-3}$, indicating that the nuisance odor could be sensed when the concentration of CH_3SH is more than 0.15 $\mu g\, m^{-3}$ in air.

TSCs are present in natural food and released into the atmosphere during the fermentation and aerobic biodegradation of food waste (Duan et al., 2014; Fang et al., 2012; Yue et al., 2014). As a result of the special dietary habits in China, MSWs in China comprise high food waste and moisture contents, which lead to serious LFG fugitive emissions and air pollution by TSCs (Qu et al., 2015).

The working faces of landfills are recognized as the major source of odor pollution in landfill sites. As fresh wastes are dumped and compacted on the working faces of landfills, they are usually exposed to air for two to three weeks before being well covered. Estimate indicates that ~20% of LFGs are diffused into the atmosphere from working faces (Spokas et al., 2006). In the present study, the emissions of TSCs, including CH_3SH, dimethyl sulfide (DMS), dimethyl disulfide (DMDS), and carbon disulfide (CS_2), from the working faces of landfills are

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investigated. These TSCs are limited by the Ministry of Environmental Protection of China under the Emission Standards for Odor Pollutants (GB14554-93). Only a few studies have investigated the odor impacts of these TSCs from the working faces of landfills in China.

For the assessment of odor impacts, olfaction measurement and odor activity value (OAV) evaluation are two alternative approaches. Olfaction method is a direct sense measurement of smell. However, it is time-consuming and costly; in addition, it could not evaluate the offensive potential of specific substances (Blank, 2002). The OAV is a widely used and cost-effective method to describe odor properties and evaluate the odor contributions of each odorant. OAV is generally calculated via chemical concentration and OTV (Feilberg et al., 2010; Wu et al., 2017). Chemical concentration is a basic requirement for odor impact evaluation using OAVs.

The objective of the present study is to characterize odor pollution of in the vicinity of a landfill caused by TSCs released from the working face. Emissions data of TSCs was acquired by long-term on-site samplings using a modified wind tunnel system. The spatial concentration distributions of the TSCs were then calculated using a Gaussian dispersion model in a "normal case" scenario and a "worst case" scenario. Finally, the odorous compounds were identified on the basis of OAVs, and their odor pollution levels were evaluated. The results provide significant evidence to comprehensively understand odor pollution caused by VSCs from MSW landfills and to formulate strategies for odor control and abatement in landfill sites.

2. Material and methods

2.1. Site description

The emissions of TSCs were measured from a typical flatland landfill located in Beijing, China (40.16° N, 116.35° E). The site has an active gas extraction system with vertical wells and horizontal collectors. Approximately 3000–3500 t d^{-1} of MSW is deposited in this landfill. The working face has an area of approximately 600 m^2, where fresh wastes are dumped and compacted daily. However, the wastes on the working face are exposed to air for approximately two to three weeks before being covered well. The composition of the landfilled waste is plotted in Fig. S1 (Supporting Information). The landfill only holds urban MSWs; thus, food waste is the dominant component, accounting for approximately 65.3% of the wet weight. Organic fractions, such as food waste, plastic, paper, textile, and wood comprise 97.4% of the total landfilled waste. Few ash wastes are landfilled.

2.2. Gas sampling and analysis

The emission rates of the VSCs were measured using a wind tunnel system modified by the authors in previous work (Liu et al., 2015). The scheme of wind tunnel system was shown in Fig. S2 (Supporting Information). Nitrogen was used as the carrier gas through the wind tunnel, and the flow rate was fixed at 19 m^3 h^{-1}, which could guarantee uniform flow performance. The system was installed on the working face of the landfill, and the carrier flow was adjusted until the target flow rate was achieved. Gas samples were withdrawn from the outlet of the system with a SOC-01 sampler (Duan et al., 2014) into a 1 L multilayer foil sampling bag (Dalian Delin Gas Packing Co., Ltd.).

Sampling campaigns were conducted on the working face of the landfill on selected days from May 2014 to January 2015, which covered four seasons in Beijing. A total of 124 valid samples were obtained. The details of the sampling procedure were described by the authors in a previous study (Liu et al., 2015). Meteorological conditions during sampling were summarized in the supporting information (Table S1). To minimize excessive heating and avoid direct exposure to sunlight, opaque sampling bags and a light-proof gas sampler (SOC-01, National Key Laboratory of Odor Pollution Control of EPA-China, Tianjin) were used during sampling. Gas bags were kept in an opaque box before measurement.

The quantitative analyses of the TSCs were conducted within 24 h after collection using an Agilent Technologies 7890A gas chromatography (GC) system with a flame photometric detector (FPD) and equipped with Rtx-Sulfur 800-365-1688 column, 30 m × 0.53 mm ID × 1.0 μm, Cat# 10155, Serial 859720. Nitrogen was used as the carrier gas with a flow rate of 30 mL·min^{-1}, hydrogen was used as the fuel gas (70 mL·min^{-1}), and air was used as the oxidant gas (60 mL·min^{-1}). The GC detector temperature was set at 250 °C. The oven was programmed initially at 35 °C for 2 min, increased to 80 °C at 5 °C·min^{-1}, and finally increased to 230 °C at 40 °C·min^{-1}. The flow rate through the column was 1 mL·min^{-1} with a pressure of 0.700 psi. Retention time was used to identify analytes. The retention times of CH_3SH, DMS, and DMDS were 0.604, 0.762, and 2.284 min, respectively.

CS_2 was analyzed with HAPSITE®ER (Inficon, East Syracuse, USA), a portable gas chromatography-mass spectrometer (GC-MS) equipped with a non-polar column (100% polydimethylsiloxane; I.D. 15 m × 0.25 mm, d_f 1.0 μm). The heating program was described in a previous study (Liu et al., 2015). CS_2 was identified in terms of its retention time, target, and qualifier ions; it was quantified using the internal standard calibration procedure, including the US National Institute of Standards and Technology 98 Library and the US Environmental Protection Agency standard solution (EPA TO-15).

Four levels (i.e., 0, 10, 20, and 40 ppbv) of CH_3SH, DMS, DMDS, and CS_2 mixture standard solution diluted with pure nitrogen (Zhaoge Chem. Ltd., Beijing, China) were used as calibration standards. In all the cases, the linear fit of the dose with $R^2 > 0.99$ was achieved.

2.3. Emissions of TSCs

The emission rates of the TSCs were computed on the basis of their concentrations in the gas samples and the nitrogen flow rates blowing through the wind tunnel, as follows:

$$ER = \frac{24 \times Q \times c}{B}, \quad (1)$$

where ER is the emission rate of a TSC, μg·m^{-2}·d^{-1}; Q is the nitrogen flushing flow rate, m^3·h^{-1}, fixed at 19 m^3 h^{-1} during all the sampling campaigns in this study; c is the concentration of TSCs, μg·m^{-3}; and B is the base area of the wind tunnel, which is 0.1 m^2.

The wind blowing through the working face of a landfill frequently affects the release of TSCs, and the emission rates vary with wind speed (Hudson and Ayoko, 2008). The relationship between emission rate and wind speed was developed on the basis of boundary layer theory by Bliss et al. (1995); this relationship can be expressed as

$$ER_v = \left(\frac{v}{0.26}\right)^{0.5} ER, \quad (2)$$

where v is the wind speed, m·s^{-1}; ER_v is the TSC emission rate at wind speed v, μg·m^{-2} s^{-1}; and ER is the TSC emission rate measured by the wind tunnel system. In the current study, the carrier gas was fixed at a speed of 0.26 m s^{-1}.

Subsequently, the TSC emission flux (F) from the working face can be estimated by

$$F = ER_v \times A, \quad (3)$$

where A is the area of the working face, which is approximately 600 m^2 for the studied landfill.

2.4. Atmospheric dispersion modeling

2.4.1. Gaussian dispersion model

A Gaussian dispersion model, the core model that is widely adopted in many plume atmospheric dispersion models (Cai et al., 2015), was used to analyze TSC dispersion. Given that the working face is a fugitive